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journal or publication title	Tsukuba geoenvironmental sciences
volume	2
page range	3-8
year	2006-12-26
URL	http://hdl.handle.net/2241/101622

Tsukuba Geoenvironmental Sciences, Vol. 2, pp. 3-8, Dec. 26, 2006

Rates of chemical weathering of porous rhyolites: ten-year measurements using a weight-loss method

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Abstract

To examine the rate of weathering, field experiments using microweight-loss techniques were carried out. Two kinds of porous rhyolite forming lava domes in Kozu-shima Island were selected: (1) a younger rock (weathering period is 2.6-ka) and (2) an older rock (20-ka). Both rocks seem to have been very similar in their initial composition and structure. They have, however, different properties in the present depending on their difference in weathering period. Rock tablets of these rhyolites with a diameter of 3.5 cm and a thickness of about 1 cm were enclosed in a nylon mesh bag and placed in a soil-bedrock interface on a hillslope of central Japan for over ten years. Results show that (1) the younger rock has a lower weight loss by about 0.4-0.8 %, and the older rock has a higher weight loss of 5 to 8 %, and (2) the rates of weight loss for both rocks on the ground surface is larger than that under the saturated grus layer. The first result shows that older rock has a higher rate of chemical weathering than younger rock. This supports Oguchi et al.'s (1994, 1999) finding that the rate of change in chemical properties appears to accelerate with weathering time. The second result suggests that in addition to chemical weathering, physical weathering such as freeze-thaw weathering occurs on the ground surface.

Key words: field experiment; rock tablet; rhyolite; weathering; weathering rates

1. Introduction

To study changing rates of landforms, geomorphologists need data on rates of weathering of rocks. Only sporadic information on rates of weathering based on quantitative measurements under natural conditions is available, as summarised by Brunsden (1979) and Kukal (1990). This limitation is mainly due to difficulties in ascertaining the time of initiation and the duration of weathering in the field. Oguchi et al. (1994, 1999) examined the weathering rates during 40,000 years based on temporal change in a variety of rock properties using four dated lava domes made of porous rhyolite with a high susceptibility to weathering and concluded that rates of change in chemical properties appear to accelerate with weathering time. To examine the finding of Oguchi et al. (1994, 1999), we performed the field experiments using the microweight-loss technique for rock tablets of two rhyolites: a younger rock (weathering period is 2.6 ka) and an older rock (20 ka) (Matsukura et al., 2001). Five years of measurements revealed that older rock has a higher rate of weight loss. This result supported the Oguchi et al.'s (1994, 1999) finding. The experiment continued and we now report further findings about the lithological and environmental effects on weathering rates.

2. Test materials and their rock properties

Successive eruptions of rhyolite formed many lava domes on Kozu-shima Island located in the western Pacific, off the southeast tip of the Izu Peninsula, 180 km south of Tokyo. Rock blocks of the two rhyolites tested were taken from two lava domes: Kobe-yama and Ohsawa-yama, formed by eruptions at 2,600 and 20,000 yBP, respectively. The rocks of these domes are hereafter called 2.6-ka and 20-ka rocks, named after the ages of the formation. Oguchi et al. (1994, 1999) have pointed out that (1) both rocks had similar original properties when lava domes were formed judging from the analyses for mineral composition and porosity, and (2) deep and uniform weathering in each lava dome indicates that the present-day rock properties can be assumed to present the degree of weathering in the time elapsed since each eruption.

Table 1, showing the rock properties of both rocks, presents that the 20-ka rock has been more highly weathered than the 2.6-ka rock reflecting the longer weathering period. The mineral composition was determined using X-ray diffraction (XRD) and a polarization microscope. Both rocks include phenocrysts of quartz, feldspar and biotite in a glassy groundmass. The composition of feldspars in each rock is 15% albite, 70% anorthite and 15% orthoclase ($Ab_{15}An_{70}Or_{15}$). Clay minerals were not detected in the 2.6-ka rock, whereas mica clay minerals and kaolin minerals were found in

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Table 1 Rock properties of two rhyolites tested

	Kobe-yama (2.6 ka)	Ohsawa-yama (20 ka)
Rock forming minerals [1)]	Qtz, Or, Pl, Bt	Qtz, Or, Pl, Bt, K, M
Chemical composition, wt (%)		
SiO_2	77.27	76.07
TiO_2	0.09	0.12
Al_2O_3	11.90	11.88
$FeO+Fe_2O_3$	0.73	0.78
MnO	0.06	0.07
MgO	0.11	0.16
CaO	0.71	0.69
Na_2O	4.47	4.03
K_2O	3.07	3.38
P_2O_5	0.02	0.02
H_2O^-	0.09	0.23
H_2O^+	1.48	2.58
Total	100.00	100.01
Physical and mechanical properties		
Specific surface area (m^2/g)	0.02	2.56
Bulk density (g/cm^3)	1.69	1.58
Porosity (%)	30.1	35.3
Compressive strength, Sc, dry (kgf/cm^2)	137	59
wet (kgf/cm^2)	113	53
Tensile strength, St, dry (kgf/cm^2)	21.2	4.7
wet (kgf/cm^2)	18.5	4.0
Equotip hardness, L_{sf}, dry	599	361

1) Qtz: quartz, Or: orthoclase, Pl: plagioclase, Bt: biotite, K: kaolin minerals, M: mica clay minerals

the 20-ka rock. Both rocks have flow structure within the glassy groundmass. The SEM photographs (Fig. 1) show that many microcracks and exfoliation structures are often found in the 20-ka rocks, whereas no microcracks are found in the 2.6-ka rock. Both rocks are vesicular and porous with a high porosity of 30.1% (2.6-ka rock) and 35.3% (20-ka rock). The 2.6-ka rock has a specific surface area of less than 1 m^2/g, while the 20-ka rock has a larger value of 2.56 m^2/g.

To evaluate rock hardness of the both rocks we tried to use an Equotip hardness tester, which is based on the same principle as a Schmidt hammer but which is with much less energy released (3 N/mm for a C type probe) and a far smaller impact area (7.07 mm^2). Aoki and Matsukura (2004a, 2004b, 2005) have recently applied this device in geomorphological studies. The Equotip hardness

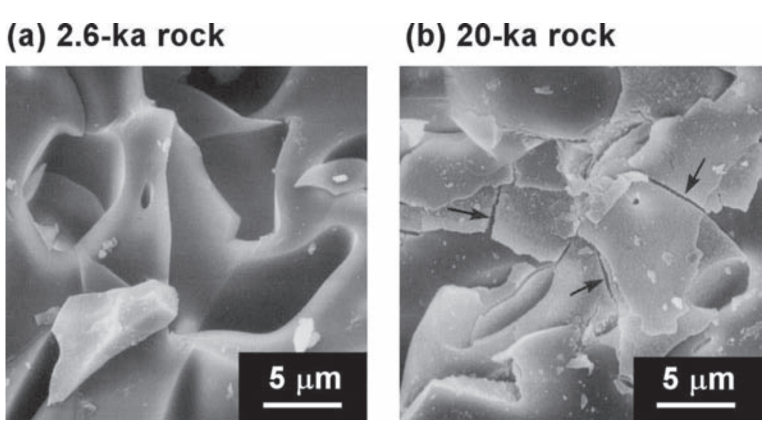

Fig. 1. SEM photographs of glass surface. Arrows in the photograph (b) for 20-ka rock show the microcracks with a width of ca. 1 μm

tester is a small electronic, battery operated device. A spherically shaped tungsten carbide test tip of diameter 3 mm is mounted in an impact body, and is driven by the spring to impact against the test surface, from which it rebounds. The impact and rebound velocities (V_1 and V_2) are measured and processed to give the hardness value L ($L = V_2/V_1 \times 1000$), which is shown on a digital display. Multiple types of impact tester (probes) are available for the Equotip. Probe C was used for all of the described tests. Aoki and Matsukura (2004b) assert that single impact testing, i.e., a single strike to various points a few mm apart from each other, is useful for evaluating the strength of a surface layer of rock. We used this technique on the unexperienced tablets and the weathered tablets after the experiments, using 30 impact points; the mean value from the five highest values of the unexperienced tablets is denoted by L_{sf}. The 2.6-ka rock has a larger L_{sf}–value of 599 than 20-ka rock (L_{sf}–value 361) (Table 1).

3. Field experiments using microweight-loss technique

3. 1. Methodology - weighing, emplacement, recovery, and reweighing

The rock blocks of both rhyolites were cored and the cores were sliced into standard-sized tablets (3.5 cm in diameter and 1.0 cm thick). We prepared five tablets for each of the two rock types. The tablets were dried at 110°C for 24 hours. After cooling, the five tablets were weighed together to an accuracy of 0.001 g using a microbalance, in order to minimise the effects of individual tablet variation. They were enclosed in a nylon mesh bag (mesh size 2.78 mm). The bags were placed in the field as described in the next section. These tablets were re-excavated every three months for the first seven years, and every six months for the last three years (total approximately ten years from the end of 1992 to February 2003). In the laboratory, the tablets were rinsed carefully with water, dried at 110°C for 24 hours, cooled and then reweighed. Changes in the mass of each sample set were monitored by weighing to ±0.001 g.

3. 2. Environmental conditions at field experiment sites

For the field experiment we used a small catchment plot of forested hillslope in the Abukuma Mountains of central Japan (37°18'53"N and 140°40'45"E: see Matsukura et al., 2001, Fig. 2). The catchment has an area of 4.1×10^{-2} km^2 (4.1 ha) and an altitude of 740 to 590 m, with an average slope angle of about 30 degrees. It is covered with Japanese cedar and broadleaf trees and is underlain with Jurassic granodiorite covered with regolith and grus, having a thickness of about 1-2 m. Regolith and grus derived from weathering of granodiorite consists of a clay size (<2 μm) fraction of 5 %, a silt size (2-63 μm) fraction of 14 %, and a sand size (>63 μm) fraction of 81 % (Terada et al., 1994). The hydraulic conductivity of the regolith is of the order of 10^{-5} m/sec.

The bags of tablets were buried in the plot at the centre of the catchment corresponding to the seepage faces (see Matsukura et al., 2001, Fig. 2). To assess the effect of environment on weathering rates, the samples were exposed to four distinct environmental conditions: on the ground surface, and buried into the humus-soil layer (15 cm deep), the soil-unsaturated grus layer (60 cm in depth) and the saturated grus layer (25 cm in depth) below the groundwater level.

Meteorological data from the Ono-nii-machi AMeDAS Station, located 6 km southwest of the experimental site, show that for the decade from 1993 to 2002 the mean annual temperature was −0.6°C in the coldest month (January), 22.9°C in the warmest month (August), and 10.6°C overall throughout the year. The average annual precipitation was 1,216 mm (pH~5.5), of which a large proportion fell between June and September. On average, there are about 120-125 freeze-thaw days annually (Suzuki, 1966). Soil freezing to about 10 cm depth was observed at the experimental site at the end of February 1994.

Groundwater (filtered to 0.22 μm) was collected in September, December and April (2004-2005), and analyzed using the ICP-AES (SPS7700, Seiko Instruments). The water temperatures were 7.7-14.0°C

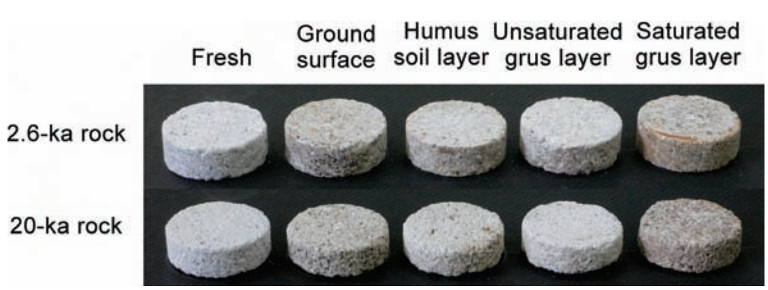

Fig. 2. Weathering features of starting specimens and those of tablets after ten-year experiments.

and pH 6-7, and the concentrations of dissolved elements (ppm) were Si 7.5-7.9, Ca 4.6-4.9, Na 2.8-4.0, K 1.1-1.3, Mg 0.78-0.85, Al 0.026-0.074, Fe(II)+Fe(III)<0.011 (Yokoyama and Matsukura, 2006). This suggests that all minerals can dissolve continuously in the groundwater.

4. Results and discussion

4. 1. Weathering features and change in hardness

Figure 2 shows an example of the starting tablets and the weathered tablets after 10 years field experiment for both rhyolites. The 2.6-ka rock was moderately weathered, while there is a clear change in the features of the 20-ka rock, in that (1) the surface of their tablets become rough and dirty, and (2) the edge of the tablets have rounded.

Hardness was measured for the weathered tablets after the experiments using the Equotip hardness tester. The method used was the same as for the fresh tablets: single impact testing was carried out 30 points on each tablet and the mean of the five highest values was taken, and denoted by L_{sw}. Figure 3 shows the results in comparison to the L_{sf} –values. The 2.6-ka rock, whatever the burial conditions, showed a large reduction of L_s–value of ca 12 to 25 %, while 20-ka rock showed a small reduction of ca. 4 to 12 %. These results show that the hardness reduces rapidly at first (2.6-ka rock) and then more slowly. This pattern is consistent with the data of Kimiya (1975), Oguchi et al. (1999) and Oguchi and Matsukura (1999), who found that rock strength reduces rapidly in the initial weathering stage, and then more slowly.

4. 2. Annual weathering rates

The weight loss for five tablets of each rhyolite is expressed as a percentage of the initial weight of the tablets. Figure 4 shows the results indicating that the 20-ka rock had a larger weight loss than the 2.6-ka rock. This is supported by Fig. 2, which shows that the edge of the tablet of the 20-ka rock becomes round. The temporal rate of loss of weight for the 2.6-ka rock is roughly constant, but the rates of weight loss for the 20-ka rock in all locations appear to decline over time. The decline in the weight loss rate is thought to result from the edges of tablets being the first to weather, due to the low hardness of these rock types. As the edge weathers away, the rate of weathering reduces.

Although as mentioned, both rocks do not show a constant rate of weight loss, the mean annual rates of weight loss are calculated using the final data in Fig. 4, as shown in Table 2. The obtained values are 0.041-0.079 %/year for the 2.6-ka rock and 0.502-0.777 %/year for the 20-ka rock.

4. 3. Weathering rates under the saturated grus layer

Since weight loss under the saturated grus layer seems to be derived from water-rock interaction or chemical weathering, the rate of weight loss means the rate of chemical weathering. The above results show that the 2.6-ka rock has a lower rate of chemical weathering than the 20-ka rock which means that the older rock has a

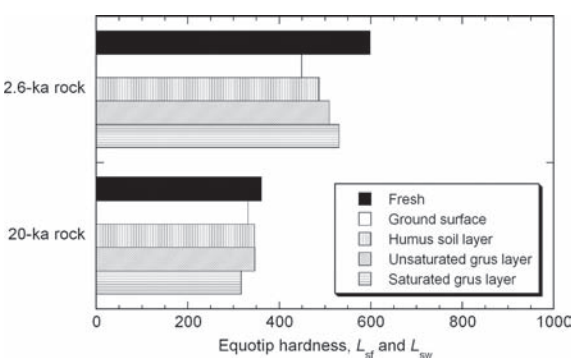

Fig. 3. Equotip hardness of tablets ("Fresh" means "unexperienced tablet"). The 2.6-ka rock has a larger reduction of L_s-value than the 20-ka rock.

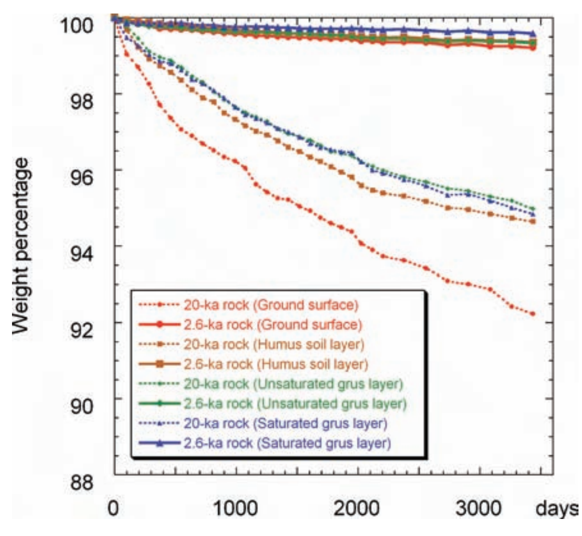

Fig. 4. Temporal changes in tablet weight loss over ten years.

Table 2 Average annual rates of weight loss calculated from the data over a ten-year period (a five-year period) (unit: % year^{-1})

Rock type	Ground surface	Humus soil layer	Unsaturated grus layer	Saturated grus layer	Mean for the four locations
2.6-ka rock	0.079 (0.110)	0.064 (0.090)	0.066 (0.094)	0.041 (0.058)	0.063 (0.088)
20-ka rock	0.777 (1.080)	0.536 (0.784)	0.502 (0.704)	0.516 (0.692)	0.583 (0.815)

higher rate of chemical weathering. This is compatible with Oguchi et al.'s (1994, 1999) finding that rates of change in chemical properties are low in the initial stage of weathering and higher in the later stages.

The reason why the older rock has a higher rate of chemical weathering is considered to be due to a higher porosity and a larger specific surface area. These characteristics, making the water-rock interaction of significant duration, result in a large amount of leaching reflected in a high rate of weight loss. The increase in porosity and surface area with increasing weathering time is derived from the following mechanism: (1) chemical weathering whereby water-rock interaction leads to hydration of the surface of glassy groundmass of the rhyolites, (2) the hydrated glass surface is warped because of expansion, which resulted in the formation of microcracks as found through SEM images (Fig. 1) and (3) further hydration occurred at a new surface of the glass by repetition of the process of (1) and (2). The feedback system of increasing surface area of water-rock interaction due to hydration accelerates the rate of chemical weathering.

4. 4. Weathering rates under the three conditions except the saturated grus layer

Weight loss under the three conditions except the saturated grus layer seems to be derived from both chemical weathering and physical weathering. There was an annual precipitation of about 1,200 mm. This percolates vertically and/or along the hillslope into the soil as subsurface flow. This water contributes to physical weathering and chemical weathering in those three environments.

Several previous studies have proposed that rates of physical weathering are controlled by rock properties such as pore properties and strength. For example, the rates of frost shattering are affected by porosity and tensile strength (e.g. Matsuoka, 1990), and growth rates of tafoni associated with salt weathering are controlled by pore size distributions and tensile strength (e.g. Matsukura and Matsuoka, 1996). These models are based on the idea that rates of weathering, i.e. susceptibility to physical weathering, is proportional to the pressure exerted by frost action or salt crystallization acting in rocks, and is in inverse proportion to the rock strength:

$$\text{Rates of physical weathering} \propto \text{pore index/rock strength}$$

Table 1 shows the data on porosity (*n*-value) and Equotip hardness values (L_{sf}–value). Since the L_{sf}–value is proportional to the compressive and tensile strength of rocks (e.g. Aoki and Matsukura, 2004a), we propose a simple model of the weathering susceptibility index (WSI-value) as n/L_{sf}.

WSI-values are calculated to be 0.079 for 2.6-ka rock and 0.111 for 20-ka rock. On the other hand, the rate of weight loss under the three environments except the saturated grus layer is 0.064-0.079 %/year for the 2.6-ka rock and 0.502-0.777 %/year for the 20-ka rock (Table 2). The relation between WSI-values and the rates of weight loss, i.e., weathering rates, shows that rocks with larger WSI-values show higher weathering rates. This suggests that in addition to chemical weathering, physical weathering occurs in these three environments. Since a freezing layer of about 10 cm depth was observed in winter, it seems probable that freeze-thaw weathering occurs as physical weathering processes on ground surface and in the soil layer. The 2.6-ka tablets buried in the unsaturated layer, which is deeper than the frozen layer, also show the same weathering rates. The cause of this is unknown.

5. Concluding remarks

A field experiment using the microweight-loss technique for rock tablets of two rhyolites revealed that older rock (the 20-ka rock) has a higher rate of weight loss (0.516 %/year) under the saturated grus layer. These results support Oguchi et al.'s (1994, 1999) finding that rates of change in chemical properties appear to accelerate with weathering time. The rates of weight loss for both rocks on the ground surface are larger than that under the saturated grus layer. These results suggest that in addition to chemical weathering, physical weathering such as freeze-thaw weathering occurs on the ground surface.

Acknowledgements

This work is partly supported by the Science Research Fund of the JSPS (16300292) through Matsukura.

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Received 31 August 2006
Accepted 28 November 2006